

# GROUNDBREAKING SCIENCE

*Experiments at the Linac Coherent Light Source aim to advance the understanding of chemistry, physics, materials science, and life itself.*

In fall 2009, researchers conducted the first experiments at the Linac Coherent Light Source (LCLS). Shown here is the Undulator Hall. (Courtesy of SLAC National Accelerator Laboratory.)



# with the WORLD'S **BRIGHTEST** X RAYS

**L**IVERMORE researchers have been among the first to use the Linac Coherent Light Source (LCLS), which produces ultrashort x-ray pulses more than a billion times brighter than ever produced on Earth. Located at the Department of Energy's SLAC National Accelerator Laboratory in Menlo Park, California, LCLS is designed to enable scientists to take stop-action pictures of atoms and molecules in motion, shedding light on the fundamental processes of chemistry, physics, materials science, electronics, medicine, and life itself.

The LCLS project is a collaboration of SLAC; Lawrence Livermore, Argonne, Brookhaven, and Los Alamos national laboratories; and the University of California (UC) at Los Angeles. Livermore experts designed and fabricated the optics that transport the x-ray beam to chambers in two experimental halls. These mirrors help control the size and direction of the x-ray beam. Additional detectors fabricated by Livermore help diagnose x-ray beam properties such as intensity.

Groundbreaking for the \$420 million facility took place in October 2006. At the dedication ceremony August 16, 2009, SLAC Director Persis Drell said, "For some disciplines, this tool will be as important to the future as the microscope has been to the past." Livermore scientists helped to characterize and troubleshoot the x-ray pulses in preparation for initial experiments in October 2009. They also led or contributed to several international collaborations that conducted some of the first scientific experiments on LCLS.





## Light Source Shines

LCLS is the world's most powerful x-ray free-electron laser (XFEL), producing pulses that are much brighter than those generated at any other x-ray laser facility (up to  $10^{12}$  photons per fleeting pulse). (See the box on p. 7.) While traditional optical lasers generate light from excited atoms, FELs use unattached, or "free," electrons moving

through a vacuum chamber at nearly the speed of light to create their beams. FEL photons are coherent, meaning they act in unison, making them well suited for imaging applications.

The laser beam's photons have wavelengths of about 0.15 nanometers, about 10 times shorter than can be produced by other XFELs, which produce "soft" or longer wavelength x rays.

A tenth of a nanometer (the diameter of a hydrogen atom) is the ideal wavelength for studying atoms and molecules and for providing new information about the atomic-level structure and dynamics of many materials.

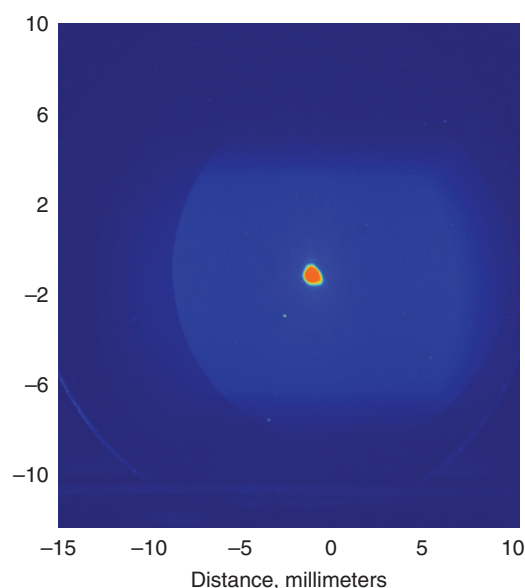
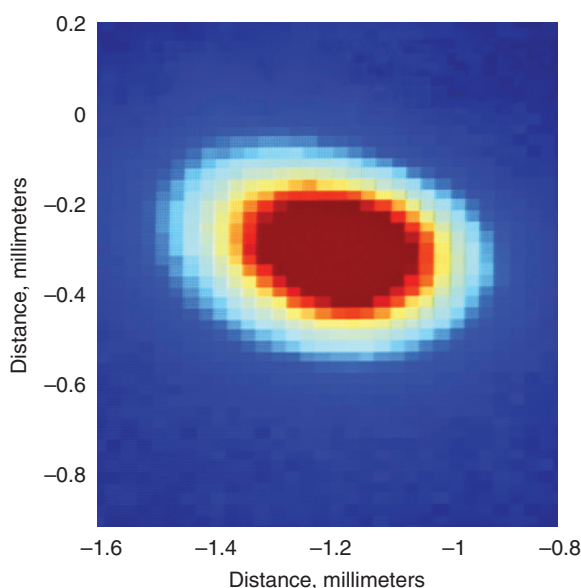
The x-ray laser pulses are of extremely short duration, lasting between a mere 10 and 100 femtoseconds (100 femtoseconds equals 100-quadrillionths of a second) and repeating 120 times per second. To put this extraordinarily short pulse in perspective, consider that light races to the moon in less than 1.3 seconds but travels just the thickness of a sheet of paper in 150 femtoseconds.

Thanks to their brightness, coherence, short wavelength, and extremely brief pulse duration, LCLS x rays will allow scientists to examine atomic-scale objects. Many fundamental processes, such as chemical and biochemical reactions, involve rearrangements of atoms and molecules that occur on timescales of femtoseconds. By sequencing separate stop-action images taken with LCLS pulses, scientists will be able to create time-resolved movies, permitting them to view chemical bonds forming and breaking in real time, such as phase transitions at the atomic level (for example, ice



Livermore scientists designed and built the LCLS x-ray transport optics and diagnostics suite. One of the suite's instruments is the x-ray free-electron laser energy monitor, which won a 2010 R&D 100 Award for being one of the year's most important industrial innovations. (See *S&TR*, October/November 2010, pp. 8–9.)

(right) An image of "first light" at LCLS in April 2009 shows an approximate 0.2-millimeter laser spot, despite having traveled more than 3 kilometers from its source. (far right) An x-ray laser pulse is viewed from the facility's main control center. A point of coherent laser light is surrounded by dimmer noncoherent light. (Courtesy of SLAC National Accelerator Laboratory.)



becoming water). Researchers also expect to create three-dimensional holograms of biomolecules.

In another type of LCLS experiment, some of the x rays scatter when they hit the sample. These deflected x rays strike a detector, and then scientists examine the pattern of diffraction. Although the powerful beam destroys each sample, the ultrashort pulse generates diffraction data before that happens. Livermore researchers have accumulated and integrated thousands of diffraction patterns to develop algorithms for converting the data into usable images.

### Collaborators Qualify the Beam

“The facility has been running very well, almost from the start,” says physicist Stefan Hau-Riege, who leads one of the Livermore experimental teams. During the summer and fall of 2009, Hau-Riege helped commission the laser. Early experiments achieved an in-depth understanding of how the beam interacts with matter as well as its operating characteristics, including the exposure required to damage materials.

LCLS experiments are performed at one of six end-stations, and the initial experiments were conducted at the atomic, molecular, and optical (AMO) end-station. This end-station, which includes x-ray optics that help focus the beam, is designed to study the effects of soft x rays (wavelengths of 0.6 nanometers and above) on atoms and molecules in gases.

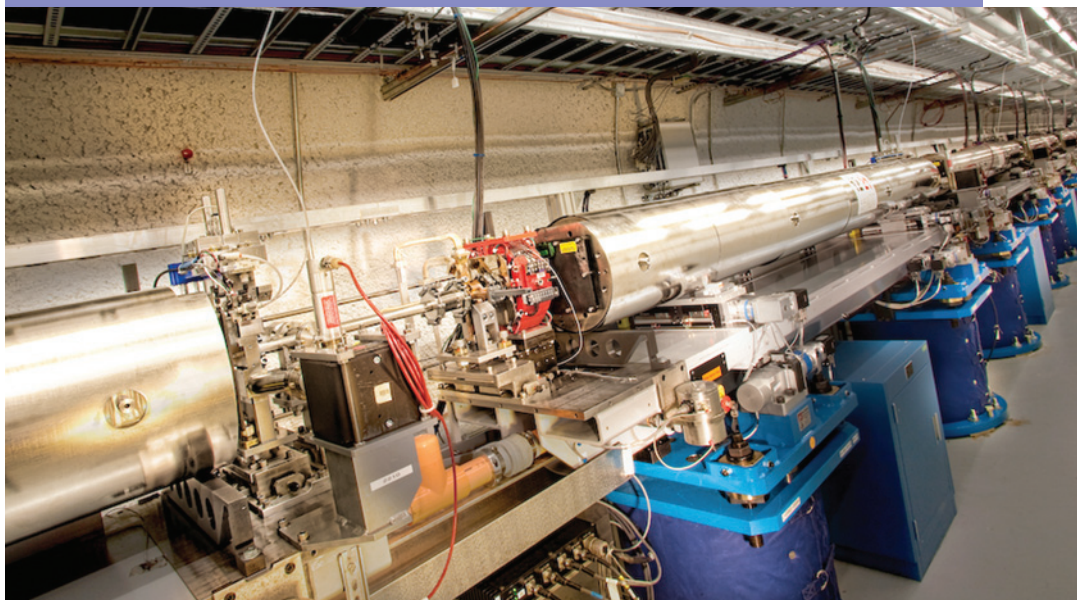
In one experiment, Hau-Riege used a 10-micrometer-diameter x-ray beam to etch the initials LCLS, about 1.5 millimeters wide, into a piece of boron carbide, a superhard substance used in accelerator shielding. (See the figure on p. 8.) To examine how the laser affects solid materials and to determine damage thresholds, the team exposed the boron carbide sample to different beam intensities and wavelengths. By moving the AMO's sample stage in minute increments, the team spelled the laser's initials in small craters blown out of the boron carbide. The

### X Rays: A Short Primer

Visible light is only a small part of the electromagnetic spectrum, which also includes radio waves, microwaves, infrared light, ultraviolet light, x rays, and gamma rays. The wavelength of light determines how small a structure we are able to see. For example, the wavelength of visible light is slightly smaller than cells and bacteria, which is why we can see them under a light microscope. Atoms have dimensions on the order of a tenth of a nanometer (a billionth of a meter), so scientists need to use a much shorter wavelength, namely short-wavelength (hard) x rays, for probing atomic structure. As a result, x rays, discovered in 1895 by German physicist Wilhelm Röntgen, have been scientists' principal means of unraveling the positions of atoms in solids from metals to proteins.

The Linac Coherent Light Source (LCLS) is the first machine of its type to probe matter with hard x rays. The facility takes up one-third of the 3-kilometer-long linear accelerator at the SLAC National Accelerator Laboratory. For more than 40 years, SLAC's linear accelerator (or linac) has produced high-energy electrons for physics experiments. Scientists are now using the linac to create x-ray pulses more than a billion times brighter than the most powerful existing sources, the so-called synchrotron sources. Synchrotrons produce streams of x-ray photons with pulses too long to explore the dynamic motion of molecules. Because LCLS can produce pulses of extremely short duration, it can achieve brightnesses significantly higher than synchrotron facilities.

SLAC's linac accelerates very short pulses of electrons to 99.99999 percent the speed of light. LCLS then takes them through a 100-meter-long stretch of alternating magnets in the Undulator Hall that force the electrons to travel back and forth. The process gives off extremely bright x rays with a wavelength of about a ten-billionth of a meter, with each pulse as short as a few quadrillionths of a second. The emitted x rays become synchronized as they interact with the electron pulses, thereby creating the world's brightest x-ray laser pulse. Each 100-femtosecond pulse, produced with a repetition frequency up to 120 hertz (currently 60 hertz), contains more than a trillion ( $10^{12}$ ) photons with energies from 500 to 10,000 electronvolts.



Extremely bright x rays are produced in the Undulator Hall of LCLS. (Courtesy of SLAC National Accelerator Laboratory.)



imprinted initials then served as a baseline for studying the effects of tests at lower x-ray energies.

To assess the depth of the x-ray-induced craters, the team used an interferometer, an instrument that measures the distance light travels as it bounces from the sample to a detector. The researchers further assessed the extent of the damage with scanning electron and atomic force microscopes.

In another experiment, the team used an XFEL energy monitor, one of three installed at LCLS, to study the interaction between nitrogen gas and x-ray pulses with energies up to 8 kiloelectronvolts. The energy monitor, developed by Hau-Riege and Livermore colleagues, measures the pulse-by-pulse energy in real time without being damaged by the beam and with minimal effect on beam quality. The total pulse energy is inferred from x-ray-induced ultraviolet fluorescent light, which is

generated by the nitrogen gas and detected by the device's photon-multiplier tubes.

"Understanding how intense x rays interact with atoms and molecules is critical to being able to take the best images and to correctly interpret the diffraction data," says Hau-Riege. The nitrogen gas experiment was performed upstream from the LCLS mirrors, which gave experimenters access to the full range of potential energies. "The sweet spot for conducting atomic-resolution imaging experiments is about 8 kiloelectronvolts," he says. At higher energies, light undergoes Compton scattering, a phenomenon where scattered x rays have different wavelengths than the incident x rays, degrading the diffraction patterns. At lower energies, sample damage is a severe problem because more of the photons are absorbed by the material. Collaborators on the nitrogen experiments included

researchers from SLAC and the Deutsches Elektronen-Synchrotron (DESY) in Hamburg, Germany.

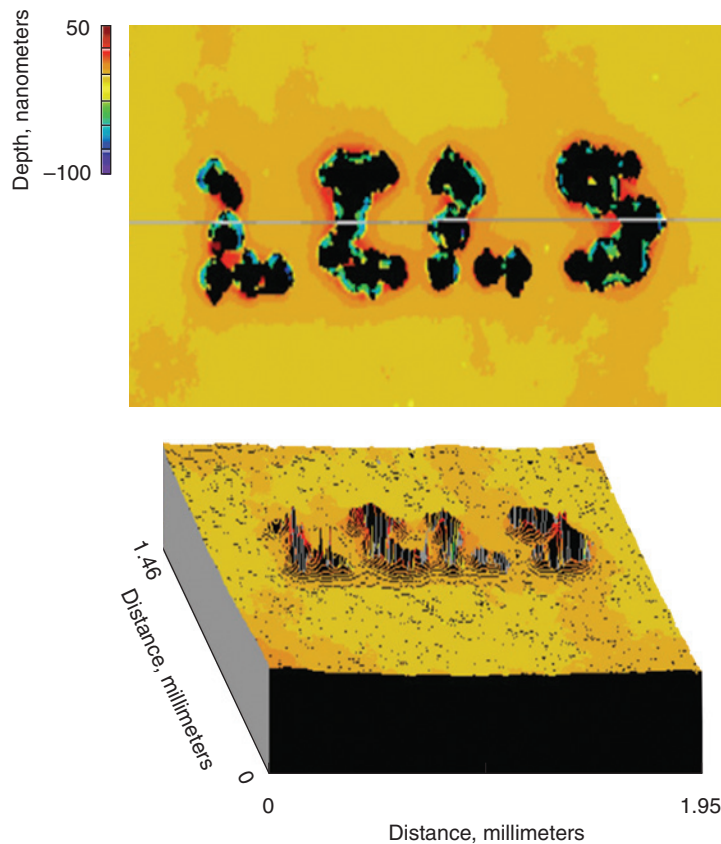
### The Real Thing

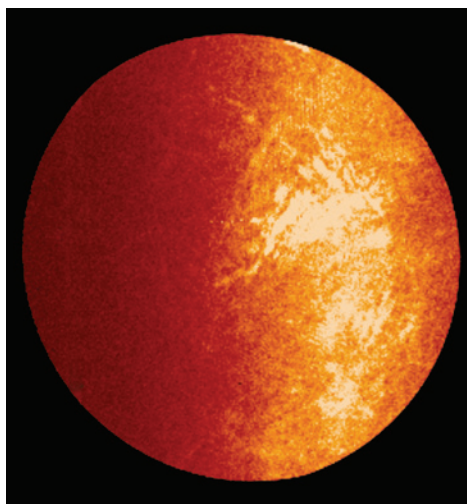
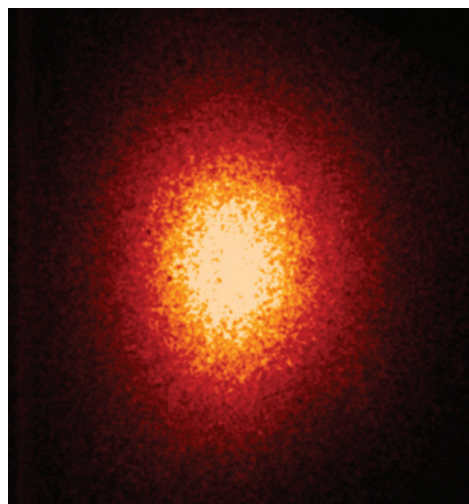
Following the beam qualification experiments, Hau-Riege led a series of experiments aimed at using LCLS to study equilibrium processes in solid-density plasmas, examine the equation of state of high-energy-density matter, and observe at near atomic scale ultrafast reactions that occur when solids are excited by x rays. The researchers imaged the microstructural lattice of graphite atoms and obtained information about ultrafast order-disorder transformations. "We saw the scattering of the x rays and measured the temperature, density, and distortion before the sample was destroyed," says Hau-Riege.

Hau-Riege explains that DOE programs in stockpile stewardship and inertial confinement fusion require high-quality data describing the properties of high-energy-density matter. In particular, data from LCLS experiments will complement those generated by the National Ignition Facility (NIF), the most energetic laser in the world, which is located at Lawrence Livermore. "NIF can create much higher pressure-density regimes than LCLS," he says. NIF, however, operates in ultraviolet wavelengths, and the "very pure" hard x rays from LCLS promise to reveal important details about excited materials to complement data from NIF experiments.

LCLS experiments to date have validated simulations Hau-Riege and other physicists have conducted using Livermore supercomputer codes. For example, LCLS results are compared with Livermore's molecular-dynamics simulation code ddcMD, which is used to model the evolution of hot, dense, radiative burning plasmas. (See the right figure on p. 9.) The experimental results demonstrate the importance of including quantum mechanical processes in these models.

Livermore researchers were among the first to commission LCLS and determine the beam exposure required to damage materials. (top) In an early experiment, researchers used a 10-micrometer-diameter x-ray beam to etch the initials LCLS, about 1.5 millimeters wide, into a piece of boron carbide. (bottom) The team then used an interferometer to assess the depth of the x-ray-induced craters. (Courtesy of SLAC National Accelerator Laboratory.)





LCLS experiments imaged the diffraction patterns of (left) single-crystal graphite and (right) nanocrystalline graphite. The single crystal measures about 1 centimeter square. The nanocrystalline graphite, of approximately the same size, is composed of many nanocrystals, each measuring about 30 nanometers in size and reconstituted from powder.

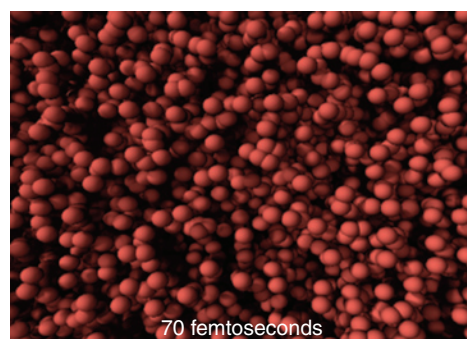
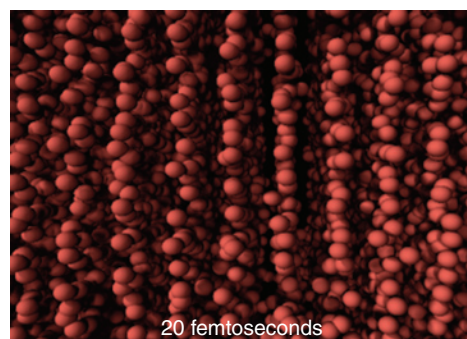
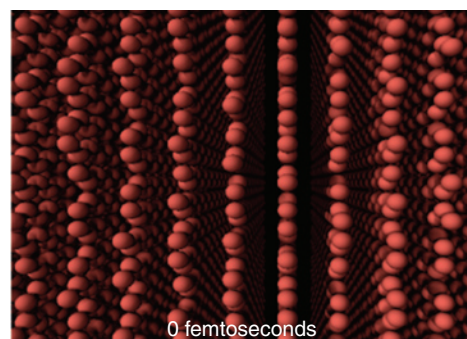
### Using One Laser to Create a Second

LCLS produces x rays through a process called self-amplified spontaneous emission, which means the x-ray beam is not temporally coherent within each extremely short pulse. Each pulse comprises 50 to 200 temporal spikes of random intensities and phases. Livermore physicist Nina Rohringer and her team have been conducting experiments to create a more temporally coherent atomic x-ray laser that is “pumped” by the LCLS x-ray pulse.

The goal is to use the LCLS beam to eject electrons from an atom’s inner shell—only x-ray photons can knock out inner-shell electrons. The “holes” in the inner shell are almost instantaneously replaced by electrons from the atom’s outer shell, in the process generating a pulse of hard x rays only a few femtoseconds long. In addition to producing shorter pulses, the secondary x-ray pulse is also considerably smoother than the LCLS x-ray beam. In September 2010, proof-of-principle experiments were conducted with neon gas. “With LCLS acting as a pump, we can create very

intense, ultrashort pulses,” says Rohringer. The experiments were conducted in collaboration with Livermore’s Hau-Riege, Jim Dunn, Richard London, Felicie Albert, Alex Graf, and Randy Hill; Colorado State University’s Jorge Rocca, Duncan Ryan, and Mike Purvis; SLAC’s John Bozek and Christoph Bostedt; and an LCLS support team.

This type of experiment was first conceived in the 1960s, but x-ray pumping sources were far too weak. In 1984, Livermore physicists developed the first atomic x-ray laser, which was based on powerful optical lasers producing collisions of electrons in hot plasmas. Rohringer says that several scientific communities might benefit from shorter x-ray pulses and improved temporal coherence. For example, shorter pulses could enable studies of chemical reactions or phase transitions in solids, while the longer coherence time would make possible, for the first time, the direct study of nonlinear quantum optics. Broadly applied nonlinear spectroscopic techniques, such as Raman spectroscopy, could be transferred from the optical



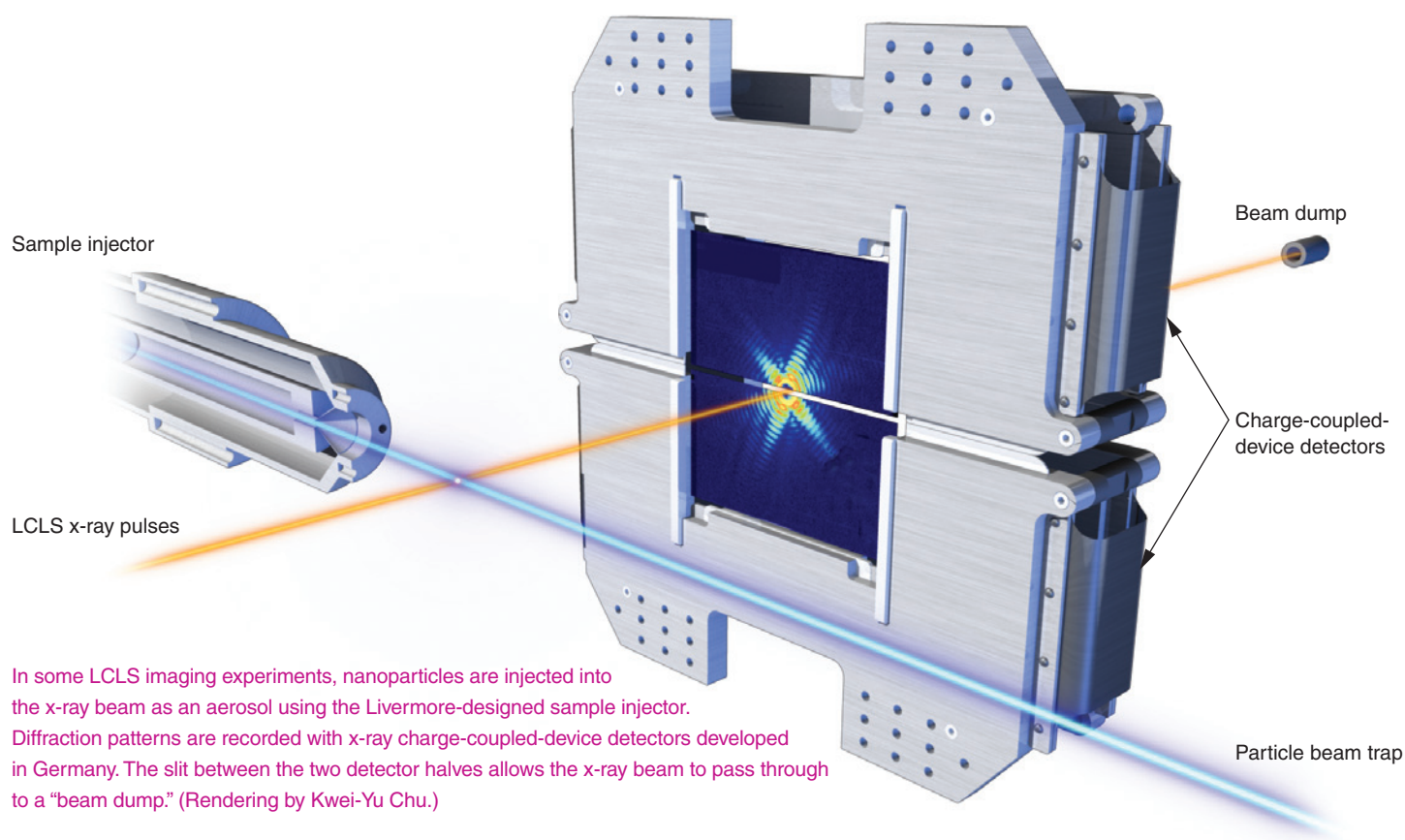
Supercomputer simulations guide Livermore experiments on LCLS. In this example, a simulated graphite lattice is shown 20 and 70 femtoseconds after exposure to the laser beam. Experiments on LCLS are largely validating the simulations.

to the x-ray regime and could result in new, powerful analytical tools to study electronic excitations in solids and molecules in a time-resolved manner.

### Imaging Biomolecules

One of the most promising avenues of LCLS research is imaging biomolecules,





including proteins, DNA, RNA, and viruses. Much of what scientists know about the detailed structure of biomolecules has come from x-ray diffraction studies of crystalline forms. Researchers traditionally have generated three-dimensional images of large biomolecules with x-ray crystallography, using x rays much weaker than those generated by LCLS. For example, x-ray crystallography was used to elucidate the double-helix structure of DNA. However, at this time, some biomolecules, such as most membrane proteins, cannot be crystallized. Studying the dynamics of noncrystalline structures requires a different x-ray diffraction technique that uses brighter x rays of shorter duration and wavelengths than have been available to date at synchrotron x-ray sources. (See *S&TR*, January/February 2009, pp. 16–18.)

Scientists have long theorized that an ultrashort and extremely bright x-ray pulse could create a single diffraction pattern from a large biomolecule before the x rays destroy the sample. If so, scientists could better understand the structure of proteins without having to crystallize them first. Key challenges have included recording ultrafast, single-shot, coherent diffraction patterns of injected biomolecules with low signal-to-noise ratio and developing robust algorithms to construct images from the diffraction patterns.

Since 2005, Livermore physicist Matthias Frank has been working on x-ray laser experiments as part of a multiyear project funded by Livermore's Laboratory Directed Research and Development Program. This effort, formerly led by physicist Henry Chapman (now at DESY and the University of Hamburg), aims

to validate the idea of using short and intense x-ray pulses to capture images of biomolecules.

In 2006, scientists made history using the FLASH soft x-ray free-electron laser at DESY. A Livermore team, including Frank, Hau-Riege, and Chapman, was part of an international collaboration with colleagues from SLAC, UC Davis, and European institutes that demonstrated how XFELs can image the ultrafast dynamics of nanoscale materials and biological structures. A Livermore-developed computer algorithm re-created an image of the object based on the recorded diffraction pattern. The Livermore team conducted additional experiments at FLASH over the next three years, gaining confidence that LCLS, when it began operation, could successfully capture images of biological materials and eventually reveal protein structure.

More recently, working with researchers from DESY, Max Planck Institute, and Arizona State University, Frank and other Livermore scientists participated in experiments in which LCLS x-ray pulses, operating at 2 kiloelectronvolts and a 6-nanometer wavelength, were used to image crystalline biological materials measuring a few nanometers in diameter. Because the AMO end-station is not designed for imaging biomolecules, the German researchers brought their own experimental chamber equipped with x-ray cameras developed at the Max Planck Institute. The chamber was bolted to the back of the end-station.

According to Frank, the biological imaging technique is revealing the structure of proteins and determining how they interact with other biomolecules. (See *S&TR*, May 2007, pp. 21–23.) Proteins are complex macromolecules that range from 400 to 27,000 amino acids in length, and their structures are a tangle of folds and twists. These structures may shift several times during their normal biological functions. Determining a protein's three-dimensional structure provides important clues about its behavior and function and guides the development of new drugs. While x-ray crystallography has revealed many secrets of protein structure, data from LCLS experiments with single molecules promise to reveal many more. UC is currently funding Frank and collaborators from UC Davis to apply x-ray imaging at LCLS to several biological molecules of interest.

Frank notes that experimentalists at LCLS will soon have the option of triggering an optical laser to fire at a sample shortly before an x-ray pulse with an adjustable time delay. Such a pump-probe technique could prove valuable as a way to determine if a protein changes shape when exposed to light.

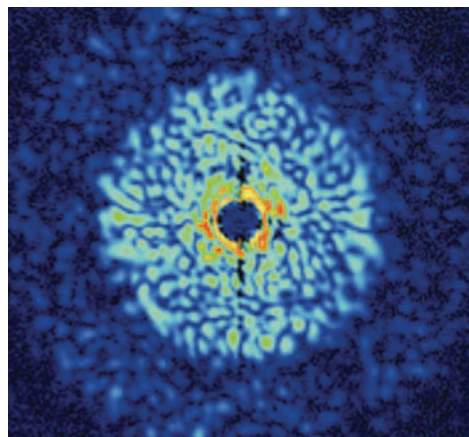
SLAC funded Frank to help develop a particle injector for the coherent x-ray imaging instrument. The instrument, which was commissioned in November 2010, is designed to image single nanoparticles, such as biological macromolecules, that are not in crystalline form. "We need to deliver nanoparticles or macromolecules from a sample into the LCLS vacuum chamber at a high rate and into exactly the right place," he says. Frank and coworkers developed an injector based on a design previously used at Livermore for single-particle aerosol mass spectrometry, which sucks ambient particles such as bacteria and viruses into a mass spectrometer to identify single particles for biodefense and environmental monitoring.

The new sample injector produces a tightly focused stream of nanoparticles aimed into the path of the incoming x-ray pulses, which hit particles on the fly. X rays scattering off atoms in the sample

particle produce a diffraction pattern revealing the particle's structure. Because the incoming x-ray pulse is so short, this diffraction pattern is produced just before the molecule explodes from the energy absorbed. The procedure is repeated many times a minute. Diffraction patterns from several individual particles can be measured each second, and thousands to hundreds of thousands of patterns are recorded in a single experiment. Even with the same type of molecule, the diffraction patterns differ because atoms and molecules are in constant motion and face the beam in different orientations. A single three-dimensional image of a representative molecule is constructed from terabytes of data consisting of many thousands of two-dimensional diffraction patterns.

At LCLS, two experimental halls will eventually house six chambers for research in atomic-molecular physics, pump-probe dynamics of materials and chemical processes, x-ray imaging of clusters and complex molecules, and plasma physics. Hau-Riege says the light source is working with little downtime compared to other facilities during their startup phase. "It is extremely stable, and the beam is easy to align." He notes that it has become highly competitive to obtain experimental time on the machine as word spreads among the scientific community of LCLS's extraordinary x-ray beam qualities.

—Arnie Heller



An experiment at the FLASH soft x-ray free-electron laser at Deutsches Elektronen-Synchrotron in Hamburg, Germany, generated an x-ray diffraction pattern from a single 1-micrometer soot particle. Similar experiments are being performed on LCLS.

**Key Words:** FLASH laser, Linac Coherent Light Source (LCLS), nanoparticles, proteins, self-amplified spontaneous emission, single-particle aerosol mass spectrometry, SLAC National Accelerator Laboratory, x-ray free-electron laser (XFEL).

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